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Supplementary Information

Ion-Exchange Strategy to Eliminate Anion Impurities within LDH Structure for Refined NCA Cathode Material

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Calculation of the apparent lithium-ion diffusion coefficient

The apparent lithium-ion diffusion coefficient of our electrodes was calculated according to the Randles-Sevcik equation:

$$I = (2.69 \times 10^5) n^{3/2} A D_{Li}^{1/2} C_{Li} v^{1/2}$$

Where *I* is the peak current, *n* is the number of electrons participating in the redox reactions, *A* is the area of the electrode, *D* is the Li^+ diffusion coefficient, *C* is the concentration of lithium, and *v* is the scan rate. The peak current (*I*) is linearly proportional to the scan rate (*v*).

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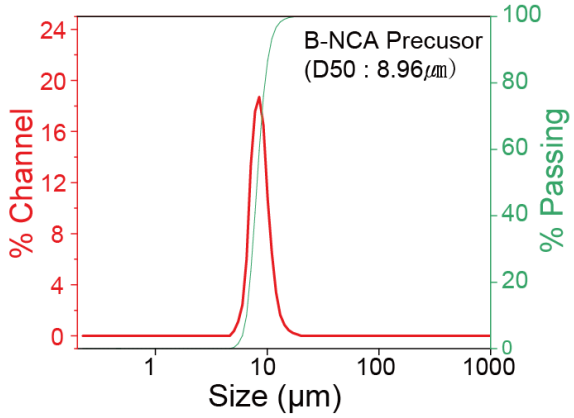

Fig. S1. Particle size distribution of Bare-NCA.

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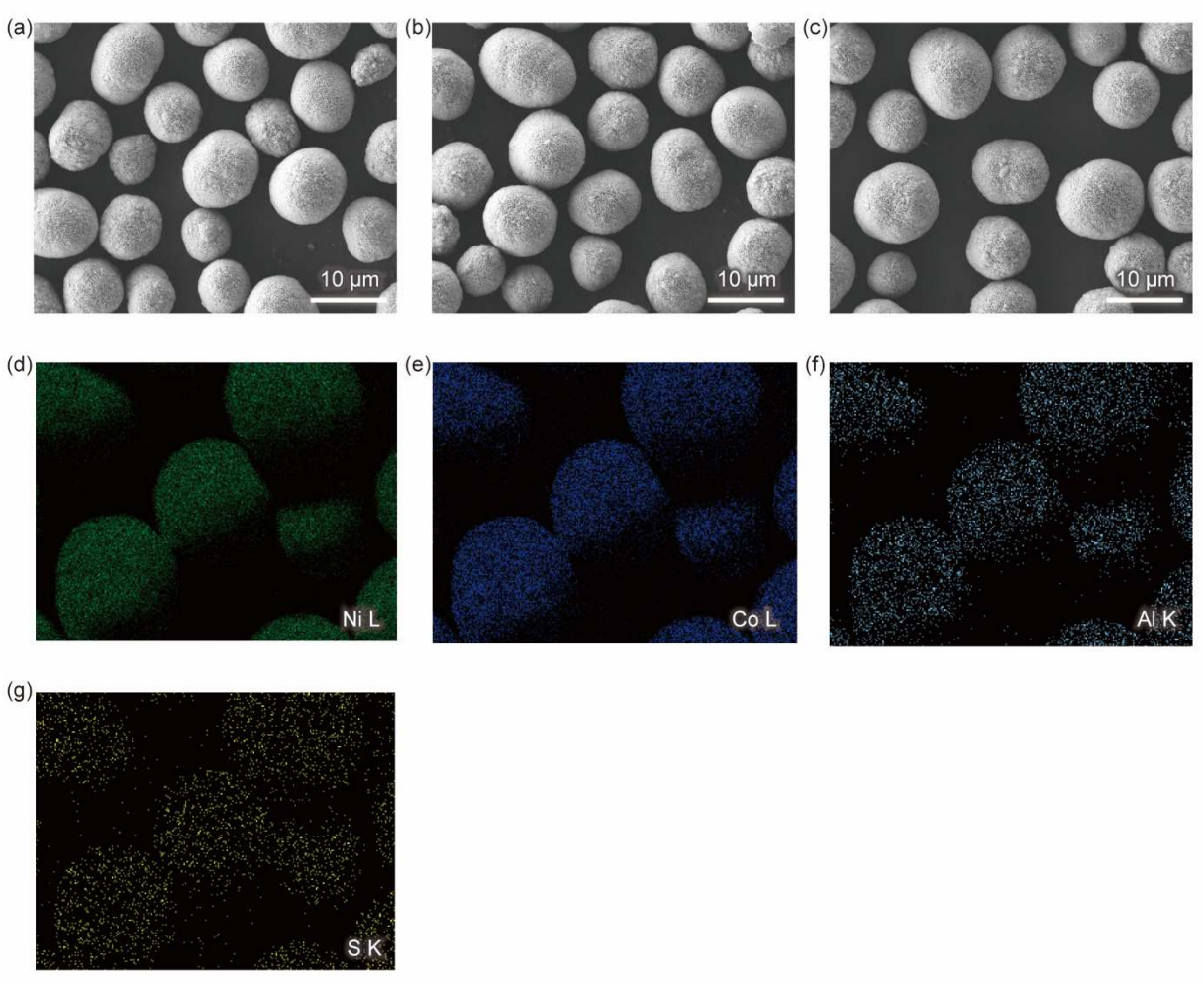

Fig. S2. SEM image of $NCA(OH)_2$ precursor (a) Bare-NCA, (b) HW-NCA, (c) Cl-NCA. EDS mapping patterns of the metal element for Bare-NCA (d-g).

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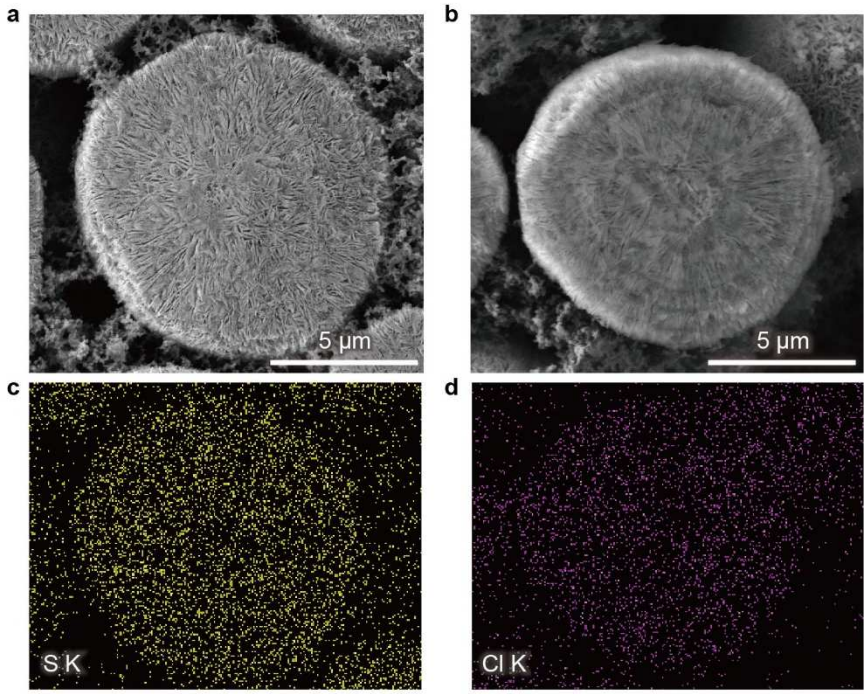

Fig. S3. Cross-section SEM image of NCA precursor (a) Bare-NCA, (b) Cl-NCA. EDS mapping patterns of the metal element for (c) Bare-NCA, (d) Cl-NCA.

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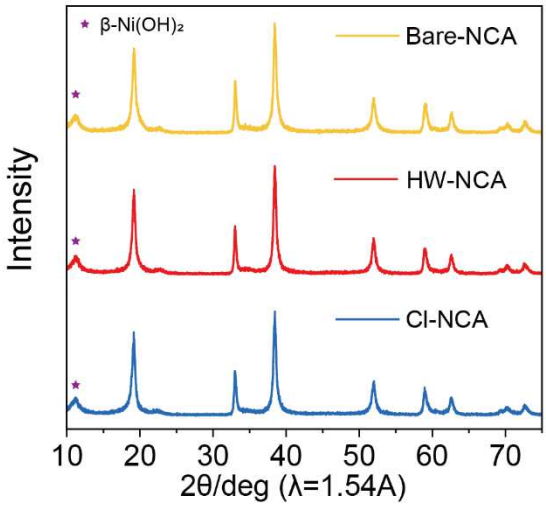

Fig. S4. XRD diffraction patterns of NCA precursors.

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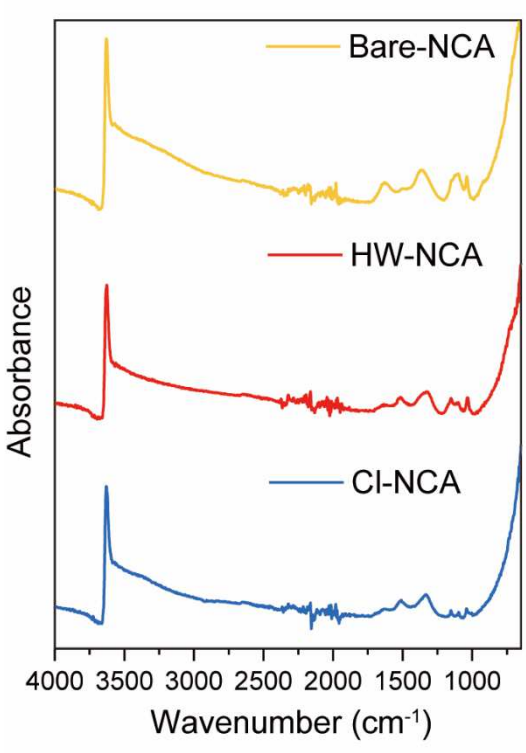

Fig. S5. FTIR spectra of NCA precursors taken in the range of 600–4000 cm^{-1}.

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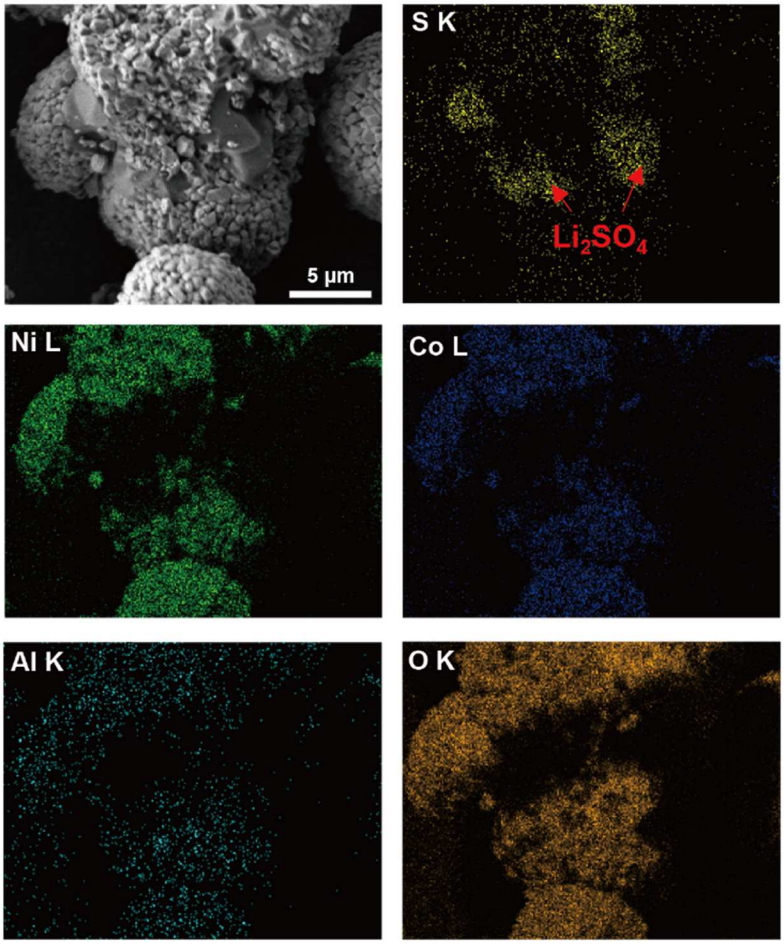

Fig. S6. EDS mapping the metal elements for Bare-LNCA.

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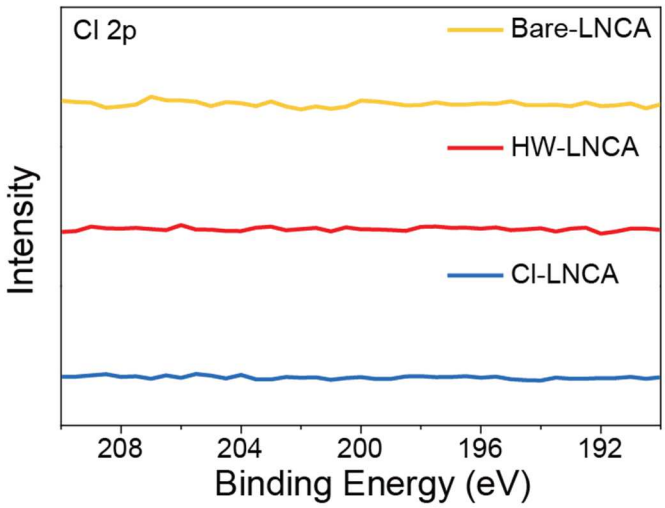

Fig. S7. XPS spectra of Cl 2p

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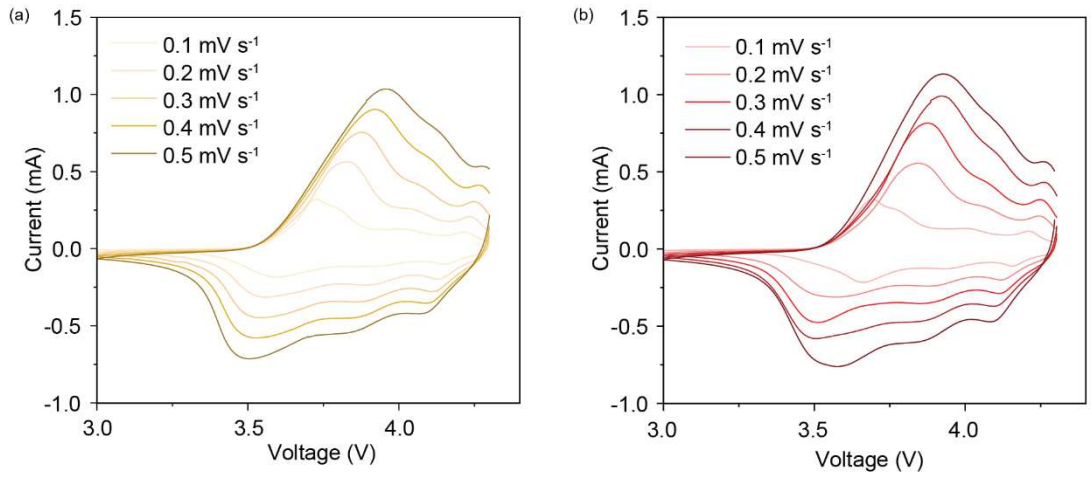

Fig. S8. Cyclic voltammetry profiles of (a) Bare-LNCA and (b) HW-LNCA at various scan rates (from 0.1 to 0.5 mV s^{-1}).

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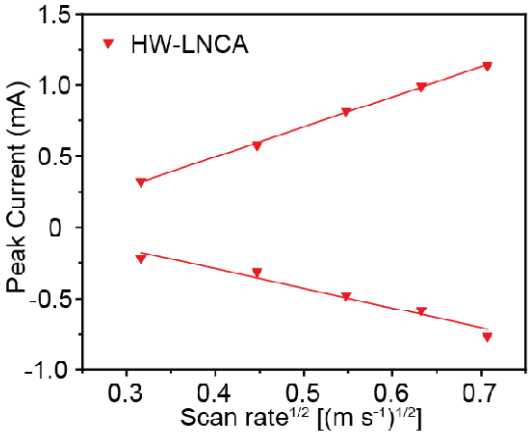

Fig. S9. Plot of the peak current vs. scan rate for HW-LNCA

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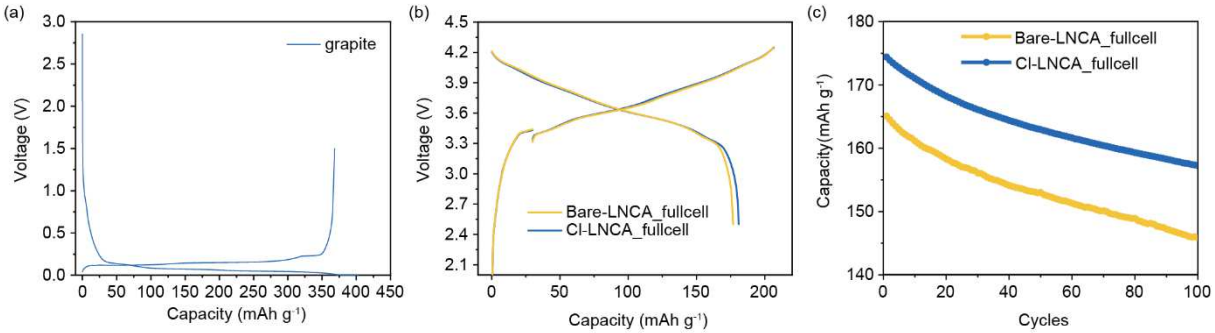

Fig. S10. (a) First charge/discharge curves of graphite within the voltage range of 0.05-1.5V (vs. Li/Li^+) at 25℃ (b) First charge/discharge curves of NCA/graphite full-cell within the voltage range of 2.5-4.25V at 25℃ and (c) Cycling performance 100 cycles at 1.0C

To facilitate sufficient electrolyte wetting and the degassing process in the full-cell, a rest period of 6 hours was applied at SOC 15% during the formation stage, followed by continued charging.

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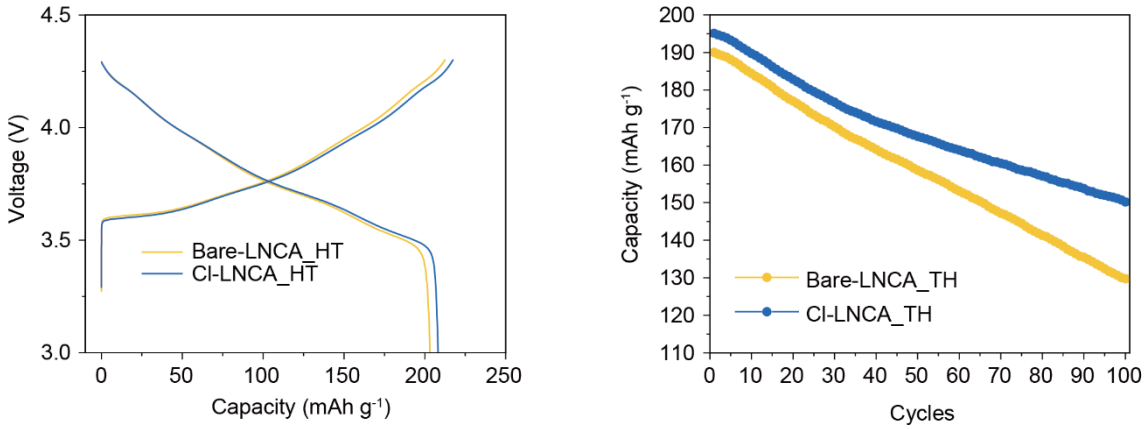

Fig. S11. (a) First charge/discharge curves of Bare-LNCA, Cl-LNCA within the voltage range of 3.0-4.3V (vs. Li/Li^+) at 45℃. (b) Cycling performance of 150 cycles for each electrode

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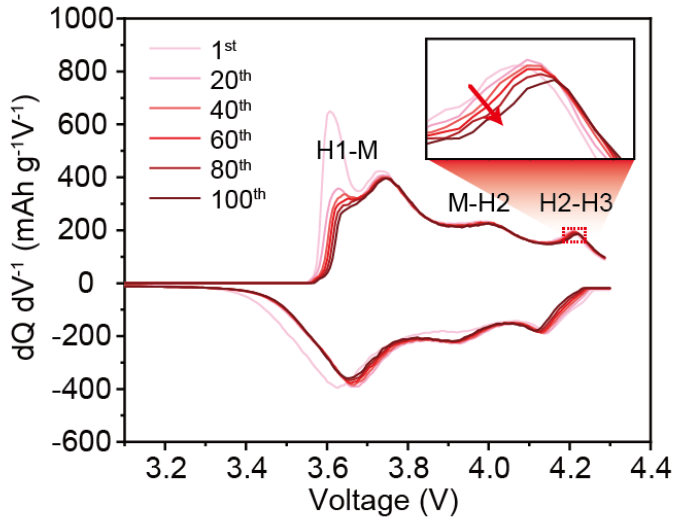

Fig. S12. dQ dV^{-1} plot of HW-LNCA at 1 to 100 cycles

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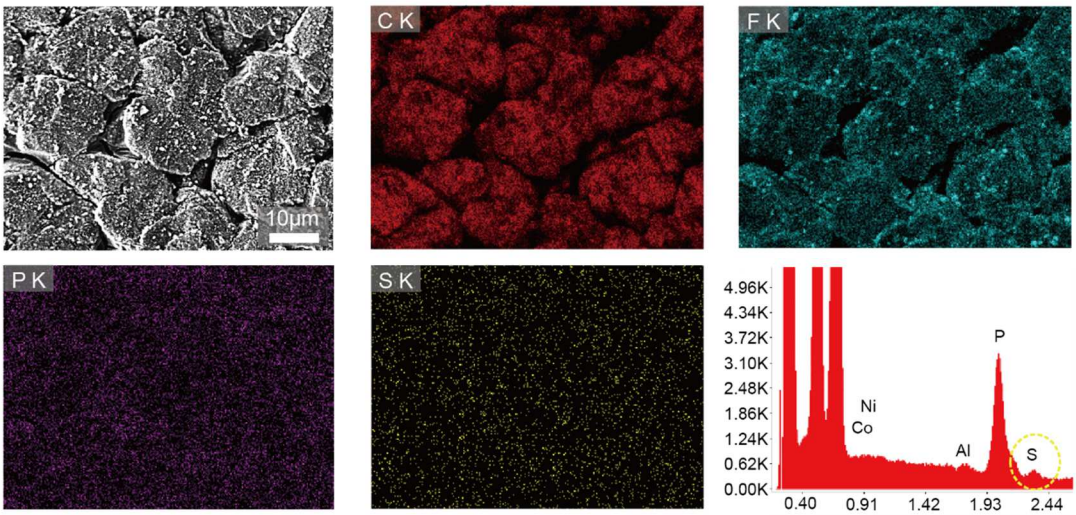

Fig. S13. EDS mapping the elements for the anode of cycled Bare-LNCA.

To confirm the surface deposition of the anode at NCA/Graphite after 100 cycles, EDS measurements of the anode electrode were performed after 100 cycles. Li_2SO_4 of the cathode was decomposed, and sulfur was measured on the anode surface.

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Table S1. Elemental composition measured by ICP Data of Bare-NCA and NaOH washed NCA precursor.

Sample	Atomic concentration (mol%)			S / TM (%)
	Ni	Co	Al	
Bare-NCA	80.0	15.5	4.6	1.72
NaOH washed NCA	77.6	18.7	3.6	0.7

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Table S2. Elemental composition measured by XRF Data of Bare-NCA, HW-NCA, and Cl-NCA.

Sample	Atomic concentration (mol%)			Ratio of transition metal (mol%)	
	Ni	Co	Al	S / TM	Cl / TM
Bare-NCA	79.1	15.6	5.3	1.93	0.0
HW-NCA	78.6	16.9	4.5	1.11	0
Cl-NCA	75.3	18.7	6.0	0.42	4.19

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Table S3. Lattice constants of the LNCA from XRD Rietveld refinement.

	a	C	c/a	R_{wp}	Cation Mixing (%)	$I_{(003)}/I_{(104)}$
Bare-LNCA	2.86453	14.1836	4.951	4.74	4.9	1.144
HW-LNCA	2.86443	14.1818	4.951	4.91	2.68	1.156
Cl-LNCA	2.86468	14.1895	4.953	4.97	2.1	1.216

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Table S4. Elemental composition measured by ICP Data of Bare-LNCA, HW-LNCA, and Cl-LNCA.

Sample	Atomic concentration (mol%)			S (ppm)
	Ni	Co	Al	
Bare-LNCA	79.7	15.3	5.0	1.15
HW-LNCA	80.0	15.0	5.0	0.97
Cl-LNCA	79.9	15.3	4.8	0.40